

Granular segregation as a critical phenomenon

Pedro M. Reis and Tom Mullin
 Manchester Center for Nonlinear Dynamics,
 University of Manchester, Oxford Road,
 Manchester, M13 9PL, UK
 (Dated: February 1, 2008)

We present the results of an experimental study of patterned segregation in a horizontally shaken shallow layer of a binary mixture of dry particles. An order parameter for the segregated structures is defined and the effect of the variation of the combined filling fraction, C , of the mixture on the observed pattern formation is systematically studied. The surprising result is that there is a critical event associated with the onset of the pattern, at $C = 0.647 \pm 0.049$, which has the characteristics of a second order phase transition, including critical slowing down.

PACS numbers: 01.50Pa, 45.70Mg, 64.75.+g, 68.35.Rh, 45.70Qj.

Keywords: Experiment, granular, binary mixture, segregation, critical phenomena, pattern formation.

Segregation is a counter-intuitive phenomenon where an initially mixed state of dry granular particles separates into its constituent components under excitation[1]. Intriguingly, it does not always happen and the conditions for its occurrence are difficult to predict. Segregation is not only of fundamental interest but it is also of practical importance with applications in areas ranging from industry[2] to geology[3]. In recent years, there has been an upsurge of interest in small scale laboratory studies where vibration [4, 5], avalanching in partially filled horizontal rotating drums [6, 7] and stratification in vertically poured mixtures [8] have all provided interesting examples of pattern formation.

Our focus is on quasi-2-dimensional horizontally driven layers of binary mixtures of particles [9, 10] as this gives the practical advantage that any collective behavior is readily visualized and gravity is effectively eliminated. Moreover, the material is in contact with the drive throughout the motion. For this class of binary granular systems a qualitative segregation mechanism has been suggested [11] borrowing the idea of *excluded volume depletion* from colloidal systems and binary alloys [12]. In a driven 2D binary system of particles of different sizes, the packing fraction of the system is decreased if self-organized clustering of the larger particles occurs. Thereby considerably increasing the number of configurational states of the system. Hence ordered arrangement of the large spheres can increase the total entropy of the system by increasing the mobility of the small particles. This process has been referred to as *entropic ordering* [1]. We believe that analogous attractive depletion forces are responsible for segregation in our granular system. We show that segregation undergoes a phase transition and occurs only for filling fractions above a critical value with the characteristics of a second order phase transition with critical slowing down.

A schematic diagram of the top view of the apparatus is presented in Fig. 1. It consisted of a horizontal smooth rectangular tray, of dimensions $(x, y) = 180 \times 90mm$ with

a flatness of less than $\pm 5\mu m$, on which a binary mixture of particles was vibrated longitudinally. The tray, made out of aluminum tool plate, was mounted on a system of four high precision linear bearings and connected to a Ling electro-mechanical shaker with a servo feedback control. The dynamical displacement and acceleration were monitored by a linearly variable differential transducer (LVDT) and two orthogonal piezoelectric accelerometers. The motion was checked to be unidirectional and sinusoidal to better than 0.1%. The forcing parameters were kept constant at amplitude $A = \pm(1.74 \pm 0.01)mm$ and frequency of oscillation $\omega = 12Hz$. Static charging effects were eliminated by coating the container's surface with a layer of colloidal graphite therefore making it conducting. Furthermore, provision was made to evacuate the apparatus and this had no influence on the segregated structures.

The binary mixture consisted of poppy seeds ($\rho_{ps} = 0.20gcm^{-3}$), which were approximately ellipsoidal with major and minor axes $d_{ps}^+ = 1.07mm$ and $d_{ps}^- \approx d_{ps}^+ \approx$

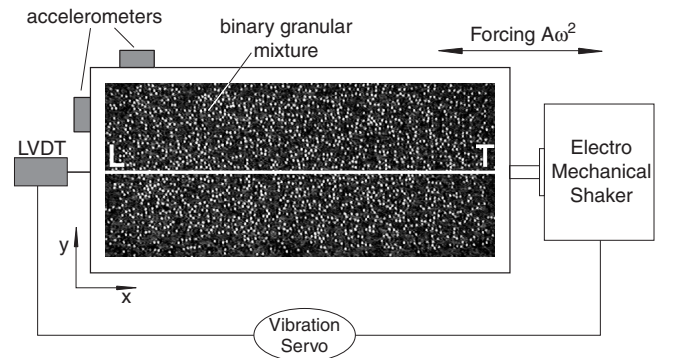


FIG. 1: Top diagrammatic view of the apparatus. The experiment is set horizontally with gravity pointing into the page. The inset photograph shows an example of the homogeneously mixed initial conditions of the granular layer with phosphor-bronze spheres (White regions = 1) and poppy seeds (Black regions = 0).

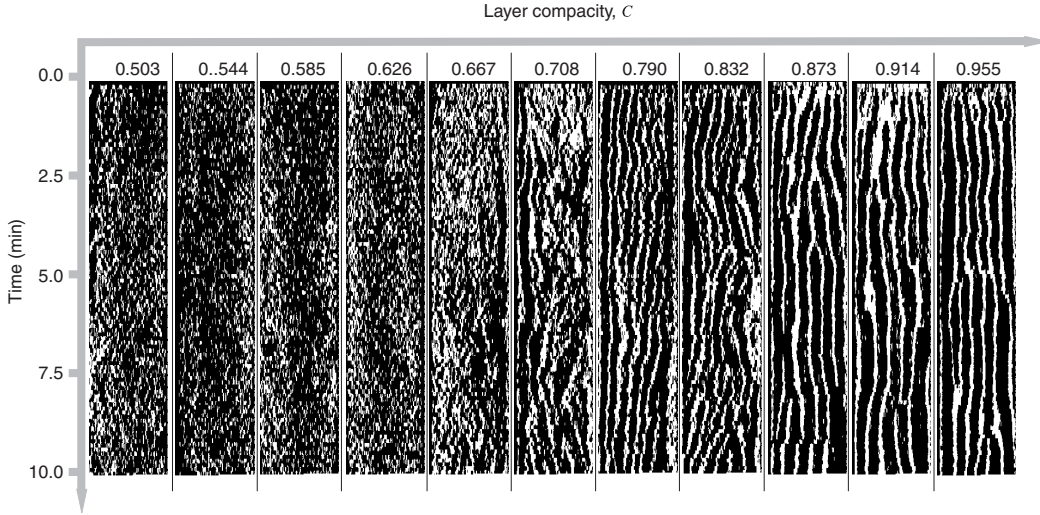


FIG. 2: Series of space-time diagrams for increasing \mathcal{C} . Each diagram is a stack, in time, of the mi-layer cut line (LT in Fig. 1). The LH side of the diagram corresponds to *gaseous* binary state. For larger values of \mathcal{C} , on the RH side, segregated structures form within the first minute of forcing. All runs were started from homogeneously mixed initial conditions.

0.50mm, respectively, and precision phosphor-bronze spheres ($\rho_{ps} = 2.18gcm^{-3}$) with diameter $d_{pb} = 1.50mm$. The amounts used were always such that a shallow layer regime was maintained, with layer height $h \leq 1.5mm$.

Forcing of individual particles arose principally from the frictional interaction with the base of the container and the effects of boundary walls were observed to be localized. Although the motion of the container was sinusoidal, the stick-and-slip nature of tray/particle contact induced stochastic thermalization; quasi-2-dimensional trajectories were obtained in x and y , with the characteristics of a 2D random walk [13]. The surface of the tray was accurately levelled with fine-pitch adjustable screws to within $\pm 0.0086^\circ$ and checked using a high precision engineers level. This ensured homogeneous forcing across the whole layer.

The granular layer was imaged using a CCD camera mounted directly above the apparatus and 600×300 pixels frames were digitized into a PC. Digital image processing techniques were employed to transform the acquired video frames into binary format, giving a value of 0 (black) to pixels corresponding to regions of poppy seeds and a value of 1 (white) for those of phosphor-bronze spheres. Frames were thereby analyzed as 600×300 digital matrices, allowing measurements on the dynamics of the granular patterns to be performed.

We define a measure of the combined filling fraction of the layer, which we call the *layer compacity*, to be,

$$\mathcal{C} = \frac{N_{ps}A_{ps} + N_{pb}A_{pb}}{xy}, \quad (1)$$

where N_{ps} and N_{pb} are the numbers of poppy seeds and phosphor-bronze spheres in the monolayer, $A_{ps} = 0.87 \pm 0.09mm^2$ and $A_{pb} = 1.77 \pm 0.06mm^2$ are the two

dimensional projected areas of the respective individual particles and x and y are the longitudinal and transverse dimensions of the tray. We regard \mathcal{C} as the principle parameter of the system.

A digitized image of a mixture of the two types of particles has been superposed on the schematic diagram of the apparatus in Fig. 1. It is representative of the initial conditions which were consistently set using the following method. Firstly, N_{ps} poppy seeds were vibrated at large amplitudes, $A \sim \pm 5mm$, creating an homogeneous and isotropic sub-monolayer. The phosphor-bronze spheres were then suspended, at a height $h = 15mm$ above the poppy seeds layer, on a horizontal frame with a perforated array of (57×28) holes, of $2mm$ diameter, arranged in a triangular lattice with a cell spacing of $a = 2.75mm$. A shutter was then opened and the 1596 phosphor-bronze spheres fell onto the layer of poppy seeds, creating a near homogeneous mixture of the two types of particles.

The focus of the investigation was to systematically increase \mathcal{C} , consequently decreasing the mobility of individual particles, and explore the conditions under which granular segregation took place. To achieve this, we incrementally increased the number of poppy seeds, N_{ps} , in the layer, in measured steps, with the amount of phosphor-bronze spheres, N_{pb} , held constant. We also experimented with changing the number of phosphor-bronze spheres and found that this did not qualitatively affect the main findings and the quantitative changes were found to be linearly scalable. We chose to perform the experiments by changing the numbers of poppy seeds since this provided finer steps in the control parameter, i.e. $\mathcal{C} = \mathcal{C}(N_{ps})$.

In Fig. 2 we present a series of space-time diagrams, for increasing \mathcal{C} , which were constructed by sampling the

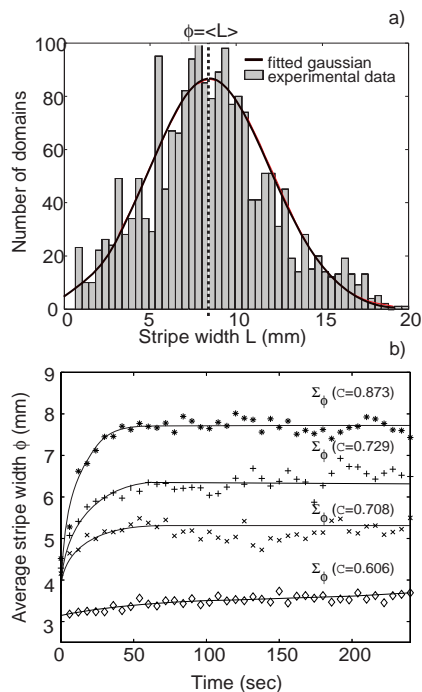


FIG. 3: a) Histogram of the L distribution (longitudinal stripe widths) for a single 600×300 pixel frame ($C = 0.966$). Solid curve is Gaussian fit. Dashed line is average value $\phi = \langle L \rangle$ which we define to be the order parameter. b) Time series for the order parameter ϕ for various values of C . A saturation level is attained after a fast initial segregation period. Solid curves are fits to Eqn. (2).

mid-frame cut line along the granular layer's x -dimension (shown in Fig. 1 as the white longitudinal line LT), and progressively stacking, in time, 200 of these (600×1 pixels) lines (acquisition rate of $1/6Hz$), for runs of 10min. This period was found to be long enough for steady state conditions to be achieved.

In the left-hand image of Fig. 2, with $C = 0.503 \pm 0.035$, it can be seen that the binary layer remains mixed for the duration of the experiment. The system acts as a low density granular gas with two components and individual particles describe quasi-2-dimensional Brownian-like trajectories. Similar behavior has been found in numerical studies of two-dimensional binary granular gases with uniform thermalization in which a mixed non-equilibrium steady state is achieved at low filling fractions [14]. The right-hand image in Fig. 2, for $C = 0.955 \pm 0.081$, displays qualitatively different behavior. It can be seen that within the first minute of the start of the motion, small localized clusters of phosphor-bronze spheres (white regions) form and progressively coalesce with neighbouring ones. Thereby coarsening occurs, so that well defined stripes are eventually formed, aligned perpendicularly to the direction of forcing [9].

At intermediate values of C , partially segregated states emerge and we aim to show that there is a critical de-

pendence of the process on the layer compacity rather than a smooth emergence of the segregated structures. Thus there is a critical value, C_c , below which the binary granular layer remains mixed. Above C_c , as the layer is incrementally compacted, segregation structures develop in a nonlinear manner. Specifically, we treat the process as if it were a second order phase transition.

In order to proceed, we first establish an order parameter, ϕ , which we choose to be the characteristic longitudinal width of the phosphor-bronze (white) stripes. For each acquired frame this was calculated by scanning through each of the 300 horizontal lines; the length of the *white steps* was measured, $L(\overset{0}{\rightarrow}\overset{1}{\leftarrow}_0)$, which typically yielded a distribution of ~ 6000 identified stripe widths. These were distributed normally, as shown in Fig. 3a, where a fitted Gaussian function has been superimposed on the experimental histogram of L . Since the distribution is well defined, we have chosen the order parameter to be $\phi = \langle L \rangle$, i.e. the average value for the distribution of longitudinal widths of the segregated stripes.

In Fig. 3b we present time series of ϕ for various C , where each run has been started from mixed initial conditions. At large compacities, e.g. $C = 0.914 \pm 0.077$, there is a fast initial growth, eventually saturating after $\sim 1min$ from the start of the experiment. At this point an approximately constant level is attained. The superimposed solid lines are fits of the form,

$$\phi^{fit}(C, t) = \Sigma_\phi(C) - \alpha \cdot \exp\left(-\frac{t}{t_s(C)}\right), \quad (2)$$

to the experimental time series of ϕ where $\Sigma_\phi(C)$, the value at which ϕ saturates, is the *segregation level*, $\Sigma_\phi(C) - \alpha$ is the initial value, $\phi(t = 0)$, and $t_s(C)$, the time scale associated with the saturation of the order parameter, is the *segregation time*.

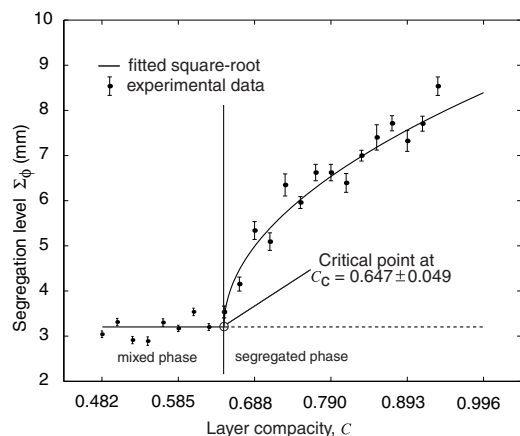


FIG. 4: Phase transition plot for segregation level. Solid line is theoretical curve for square-root singularity. Critical point occurs at $C_c = 0.647 \pm 0.049$. The mixed phase exists below this point and the segregated phase above it.

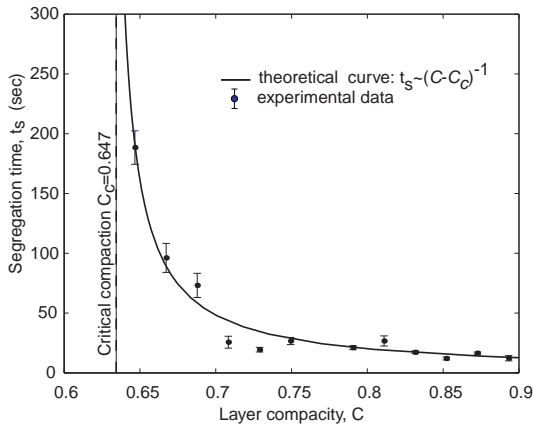


FIG. 5: Plot of segregation time, t_s , which diverges as the critical point C_c is approached from above. Solid line is theoretical curve of the form: $t_s \sim (C - C_c)^{-1}$.

The C -dependence of the segregation level is presented in Fig. 4. We see that for low compacities, up to a value, $C_c = 0.647 \pm 0.049$, the segregation level remains constant at $\langle \Sigma_\phi \rangle = 3.21 \pm 0.08 mm$. This corresponds to approximately two sphere diameters so that any domains are not classified as structures and the system is deemed to be in a mixed or unsegregated state. As C is increased past C_c , segregated stripes of increasing ϕ emerge and the segregation level exhibits a square-root singularity. The solid curve in Fig. 4 is the line of best fit of the form,

$$\Sigma_\phi^{fit} = A(C - C_c)^\beta + \Sigma_\phi(C_c), \quad (3)$$

where A is the transition scaling factor, $\{C_c, \Sigma_\phi(C_c)\}$ is the critical point at which the transition occurs and $\beta = \frac{1}{2}$ is the order parameter exponent. The numerical values for the fitted parameters were $A = 2.81 \pm 0.07 mm$, $C_c = 0.647 \pm 0.049$ and $\Sigma_\phi(C_c) = 3.21 \pm 0.08 mm$.

It is interesting to note that the generally accepted value of maximum packing fraction for a random closed packing for a 2D system of particles [15] is around 0.82 which is considerably larger than C_c for segregation in our system. This seems to indicate that the segregation transition occurs while the granular mixture is still in the monolayer regime.

A further measure that supports the notion of critical behavior is the C -dependence of the segregation time, t_s , extracted from the fittings of $\phi(C, t)$ to the function in Eqn. (2). Note that t_s is only defined for $C > C_c$, corresponding the region where segregation occurs. The measured segregation time is plotted in Fig. 5 as a function C . As expected for a second order transition, t_s diverges, as C is decreased from above, near C_c . This is usually referred to as *critical slowing down* where the divergence has the form of $t_s \sim (C - C_c)^{-\gamma}$ [16]. The solid line in Fig. 5 corresponds to the mean field approximation result, $\gamma = 1$ [17].

In summary, we have shown that granular segregation in a horizontally shaken layer displays both qualitative and quantitative features which are consistent with a second order phase transition, i.e. there is a critical layer compacity value below which mixing of the binary layer is guaranteed and above which reproducible segregation patterns form. The phenomena were not affected when the granular layer was evacuated, establishing that this is a fundamental process intrinsic to the dynamics of the granular layer. The critical exponents for the order parameter and the segregation time agree with those of mean field type behavior, $\beta = \frac{1}{2}$ and $\gamma = 1$, respectively. This is surprising since the system is far from equilibrium; it is both forced and the principal energy losses are due to inter-particle inelastic collisions. A reconciliation between this results on granular segregation and recent theoretical reports of critical behavior in granular hydrodynamics [18] and the existence of self-organized domains in granular compaction models [19] may be crucial to our further understanding of segregation and other collective phenomena in granular materials.

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